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## (54) N-(substituted cyclic alkyleneimine)-alpha-(3,5-di-alkyl-4-hydroxy-phenyl)-alpha', alpha"-dialkyl acetamides.

(57) N-(substituted cyclic alkyleneimine)-$\alpha$-(3,5-di-alkyl-4-hydroxyphenyl)-$\alpha'$,$\alpha''$-dialkyl acetamides having the general formula

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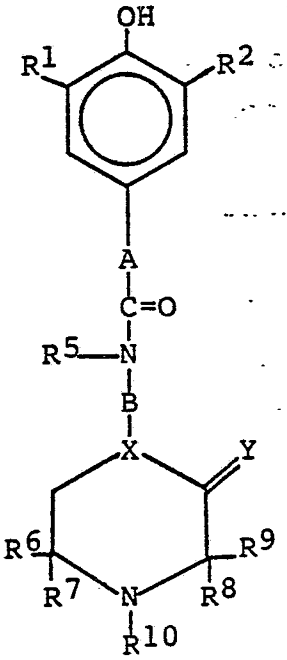


wherein $R^1$ and $R^2$ are hydrogen or alkyl and cycloalkyl groups containing 1 to 12 carbon atoms, alkylcycloalkyl and aryl or alkaryl groups wherein the alkyl groups contain 1 to 8 carbon atoms; $R^5$, $R^6$, $R^7$, $R^8$ and $R^9$ are hydrogen, alkyl and cycloalkyl groups containing 1 to 12 carbon atoms, alkylcycloalkyl and aryl or alkaryl groups wherein the alkyl groups contain 1 to 8 carbon atoms, A is, among others, an alkylidene group of the formula

$R^3\text{-}\underset{|}{\overset{|}{C}}\text{-}R^4$ wherein $R^3$ and $R^4$

are alkyl groups containing 1 to 18 carbon atoms, $C_1$-$C_{18}$; B is a bond or alkylene group $-(CH_2)_n$-wherein n is 1 to 8; $R^{10}$ is hydrogen or alkyl radicals containing 1 to 18 carbon atoms, O or OH; X is N or CH; and Y is $H_2$ or O, prepared by a new process are novel stabilizers for organic materials subject to attack the degradation, and form useful combinations for this purpose with other stabilizers against attack of polymer materials by heat, oxygen and light.

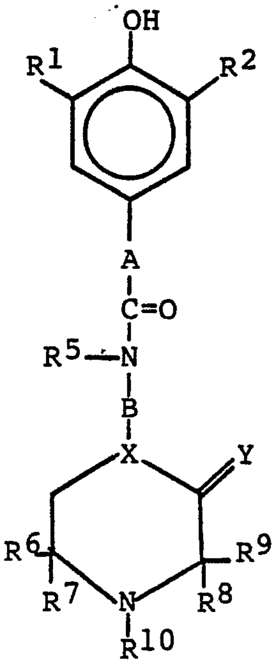
# N-(SUBSTITUTED CYCLIC ALKYLENEIMINE)-α -(3,5-DI-ALKYL-4-HYDROXYPHENYL)-α',α"-DIALKYL ACETAMIDES

## SUMMARY OF THE INVENTION

Novel N-(substituted cyclic alkyleneimine)-α -(3,5-di-alkyl-4-hydroxyphenyl)-α',α"-dialkyl acetamides prepared by a new process are novel stabilizers for organic materials subject to attack and degradation by heat, oxygen and light, to form useful combinations for this purpose with other hindered phenol stabilizers.

## DETAILED DESCRIPTION

The novel N-(substituted cyclic alkyleneimine)-α-(3,5-di-alkyl-4-hydroxyphenyl)-α',α"-dialkyl acetamides of this invention have the general formula



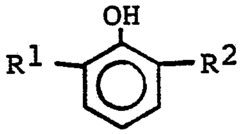
wherein $R^1$ and $R^2$ are hydrogen or alkyl and cycloalkyl groups containing 1 to 12 carbon atoms, alkylcycloalkyl and aryl to alkaryl groups wherein the alkyl groups contain 1 to 8 carbon atoms, and the aryl groups are normally phenyl or naphthyl; $R^5$, $R^6$, $R^7$, $R^8$ and $R^9$ are hydrogen, alkyl and cycloalkyl groups containing 1 to 12 carbon atoms, alkylcycloalkyl and aryl or alkaryl groups wherein the alkyl groups contain 1 to 8 carbon atoms, and the aryl groups are normally phenyl or naphthyl; A is (1) an alkylidene group of the formula

$R^3\text{-}\overset{|}{\underset{|}{C}}\text{-}R^4$ wherein $R^3$ and $R^4$

are alkyl groups containing 1 to 18 carbon atoms, $C_1$-$C_{18}$, or (2) a cycloalkyl group containing 5 to 12 carbon atoms, and alkaryl groups wherein the alkyl groups contain 1 to 8 carbon atoms, including for example those groups derived from cyclohexanone, cycloheptanone, alkyl aryl ketones wherein the alkyl groups contain 1 to 8 carbon atoms and the aryl preferably is phenyl or naphthyl, and the like; B is a bond or an alkylene group of the formula $-(CH_2)_n$ -wherein n is 1 to 8, preferably n is 2 to 4; $R^{10}$ is hydrogen or alkyl radicals containing 1 to 18 carbon atoms, O or OH; X is N or CH; and Y is $H_2$ or O. Preferably, $R^1$ is an alkyl group containing 1 to 8 carbon atoms, $C_1$-$C_8$, and $R^2$ is an alkyl group containing 1 to 5 carbon atoms, and more preferably at least one of $R^1$ or $R^2$ is a t-alkyl group including t-butyl and t-amyl; $R^3$ and $R^4$ are alkyl groups containing 1 to 8 carbon atoms, $C_1$-$C_8$, and more preferably 1 to 4 carbon atoms; $R^5$ is an alkyl group containing 1 to 6 carbon atoms; $R^6$ and $R^7$ are methyl; $R^8$ is an alkyl group containing 1 to 3 carbon atoms; $R^9$ is an alkyl group containing 1 to 4 carbon atoms; $R^{10}$ is hydrogen; in B n is 2 to 4; and X is CH.

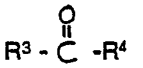
These N-(substituted cyclic alkyleneimines)-α-(3,5-di-alkyl-4-hydroxyphenyl)α′,α″-dialkyl acetamides, more specifically the N-(2,2,6,6-tetraalkyl-4-pipereidinyl)-α-(3,5-di-alkyl-4-hydroxyphenyl)-α′,α″-di-alkylacetamides and N-alkyl-N′-(3,3,5,5-tetralkyl-2-keto-1-piperazinealkyl)-α-(3,5-di-alkyl-4-hydroxyphenyl) -α′,α″-dialkylacetamides, are prepared by a new process by reacting a 2,6-di-alkylphenol with an aliphatic, cycloaliphatic or alkaryl ketone, a haloform and a piperidine or piperazine compound in the presence of alkali metal hydroxide. An organic solvent may be used, or large amounts of the ketone reactant may be employed. The amide is isolated in excellent yields by crystallizing the filtered reaction product.

Typical 2,6-dialkyl phenols used have the formula



wherein $R^1$ and $R^2$ have the meanings set forth above, including 2-methyl-6-t-butylphenol, 2-ethyl-6-t-butylphenol, 2-propyl-6-t-butylphenol, 2-isopropyl-6-t-butylphenol, 2-n-butyl-6-t-butylphenol, 2,6-di-t-butylphenol, 2-n-amyl-6-t-butylphenol, 2-isoamyl-6-t-butylphenol, 2-hexyl-6-t-butylphenol, 2-heptyl-6-t-butylphenol, 2-isooctyl-6-t-butylphenol, 2-isopropyl-6-methylphenol, 2-n-butyl-6-isopropylphenol, 2-isoamyl-6-ethylphenol, 2-isoamyl-6-methylphenol, 2-isooctyl-6-methylphenol, 2-isooctyl-6-ethylphenol, 2-isooctyl-6-n-propylphenol, 2-isooctyl-6-n-hexylphenol, and the like.

The ketones used include dialkyl ketones, cycloalkanones, alkylcycloalkanones, and alkyl aryl ketones. Typical ketones that may be used in the novel process to make the new 3,5-dialkyl-4-hydroxyphenyl-substituted acetic acids of the invention are alkyl ketones of the formula



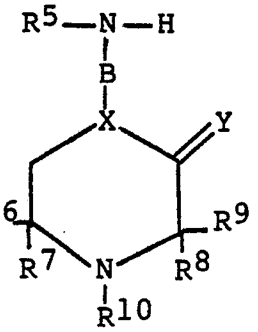
$$R^3-\overset{\overset{\displaystyle O}{\|}}{C}-R^4$$

wherein $R^3$ and $R^4$ are alkyl radicals containing 1 to 18 carbon atoms, preferably 1 to 8 carbon atoms, $C_1$-$C_8$, including for example acetone, methyl ethyl ketone, methyl n-propyl ketone, diethyl ketone, 2-hexanone, 3,hexanone, di-n-propyl ketone, 2-octanone, methyl isopropyl ketone, and the like. Also useful are the cycloalkane ketones containing 5 to 12 carbon atoms in the cyclic ring such as cyclobutanone, cyclopentanone, cyclohexanone, cycloheptanone, cyclooctanone, cyclodecanone, methylcyclopentanone, methylcyclohexanone, dicyclohexyl ketone, alkyl aryl ketones when the alkyl group contains 1 to 4 carbon atoms such as acetophenone, o-methoxyacetophenone, p-chloroacetophenone, and the like; in amounts from at least about 1 mole of ketone per mole of 2,6-dialkylphenol, up to amounts sufficient for the ketones to be the solvent of the reaction, 10 moles or more. No more than about two moles is preferred when the ketone is a reactant only. Use of less than 1 mole reduces the product yield, and more than about 2 moles is unnecessary unless the ketone is the solvent.

The haloform, such as chloroform, is also used in a molar ratio of at least about one mole per mole of 2,6-dialkylphenol used, but preferably a slight molar excess is used up to about a 50 percent molar excess, i.e., 1.5 mole per mole of 2,6-dialkyl phenol. While larger amounts may be used, there is no advantage realized, and lesser amounts will decrease the ultimate yield of the desired product. Bromoform may be substituted for the chloroform and excellent results will be obtained.

The alkali metal hydroxyide is used in powder form, or in solution, and includes sodium hydroxide and potassium hydroxide. Preferably, a molar excess of the alkali metal hydroxide is used in relation to the amount of 2,6-dialkylphenol present. Normally from about 4 moles of alkali metal hydroxide per mole of 2,6-dialkylphenol up to 10 or more moles may be used, but the amount used preferably is about 4 to about 8 moles of hydroxide per mole of 2,6-dialkylphenol. Use of less than about 4 moles will reduce the yield of desired product.

The N-substituted cyclic alkyleneimines used in the process of this invention to make the novel acetamides, the piperidines and the piperazines, have the general formula



wherein $R^5$, $R^6$, $R^7$, $R^8$ and $R^9$ are hydrogen, alkyl, cycloalkyl or alkaryl radicals containing 1 to 18 carbon atoms, more preferably wherein the alkyl groups contain 1 to 8 carbon atoms, $R^{10}$ is hydrogen, alkyl or alkaryl as defined above, oxygen or hydroxyl; X is N or CH; Y is $H_2$ or O, more preferably X is CH; B is a bond or an alkylene group of the formula $-(CH_2)_n-$wherein n is 1 to 8, preferably n is 2 to 4. Typical compounds are substituted piperidine and piperizine derivatives such as 4-amino-2,2,6,6-tetramethyl piperidine, N'-(2-isopropylaminoethyl)-3,3,5,5-tetramethyl-2-piperazinone, N'-(2-cyclohexylamino ethyl)-3,3,5,5-tetramethyl-2-piperazinone, 4-methylamino-2,2,6,6-tetramethyl piperidine, N'-(20-isobutylaminoethyl)-3,5,5-trimethyl-3-isobutyl-2-piperazinone, N,N'-(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine, and the like.

The amount of substituted piperidine or piperazine used in the reaction is based on one mole of the 2,6-dialkylphenol. While the amount may vary from less than a stoichiometric amount, resulting in low product yield, to larger excesses that are not required and have to be separated from the reaction mixture, normally about 1 to 4 moles to about 1 mole is used.

The solvent used may be any polar organic solvent, including an excess of the ketone used in place of an added solvent. Typical solvents that may be used include methylene chloride, tetrahydrofuran, diethyl ether, dibutyl ether, dimethyl sulfoxide, 1,4-dioxane, carbon tetrachloride, toluene, xylene, and the like. The amounts of solvent used will vary from about 5 moles to 100 moles per mole of 2,6-dialkylphenol used. As has been stated, the solvent may be eliminated if an excess of the reactant ketone is used. In this case, the amount of ketone used, based on the moles of 2,6-dialkylphenol used may be from about 5 to about 20, preferably about 7.5 to 15 moles per mole of 2,6-dialkylphenol.

While the reactants may be added in any order, it is preferred that the alkali metal hydroxide be added last, over a period of time to control the exothermic reaction and preferably maintain the reaction below 30°C, preferably below about 10°C. The reaction temperature may be varied from about 0°C to about 30°C, but preferably is conducted from about 0°C to 10°C. The reaction time normally will vary from about 5 to about 15 hours.

The amides are readily isolated from the reaction mixture by filtration, washing the filtrate with aqueous inorganic acid, including hydrochloric or sulfuric acid, to the reaction mixture to isolate the amide, most of which is in the organic layer that forms. The aqueous layer may be extracted with solvent to remove all traces of the amide, and this is added to the other organic layer and this mixture is dried with a desiccant such as anhydrous sodium sulfate, and heated to dryness. This product may be recrystallized if desired.

The practice of the invention is demonstrated in the following Examples. The structures of the amides of the following Examples were confirmed by infrared and nuclear magnetic resonance spectra. Molecular weights were determined and confirmed by field desorption mass spectra (FD/MS). Elemental analysis of carbon and hydrogen was done and the amounts found were consistent with the formulas of the materials.

## EXAMPLE 1

### N-methyl-N'-(2,2,6,6-tetramethyl-4-piperidinyl)-2-(3,5-di-t-butyl-4-hydroxyphenyl)-2-methyl propionamide

0.1 Mole of 2,6-di-t-butylphenol, 1.0 mole of acetone, 0.15 mole of chloroform and 0.1 mole of 4-methylamino-2,2,6,6-tetramethylpiperidine were added to a reactor and mixed by stirring while being cooled to 5°C by a circulating cold bath. 0.5 mole of powdered sodium hydroxide was slowly added in small portions over a period of 1 hour. The reaction mixture was stirred at 10°C overnight. The reaction mix from the reactor was filtered. The solid residue was washed with methylene chloride and the wash added to the filtrate. The filtrate was washed with 50 ml 4N hydrochloric acid, 50 ml of 5% sodium carbonate, and was then dried over sodium sulfate. The filtrate was evaporated to dryness and the dried product was washed with hexane. The resulting amide was a white solid that had a melting point of 168-173°C and a molecular weight of 444. By elemental analysis it was determined that the amide contained 75.38% carbon (75.67% calculated), 10.76% hydrogen (calculated 10.88%) and 6.20% nitrogen (calculated 6.30%).

## EXAMPLE 2

### N-Isopropyl-N'-[2-(3,3,5,5-tetramethyl-1-piperazinyl)]ethyl-2-(3,5-di-t-butyl-4-hydroxyphenyl)-2-methylpropionamide

0.1 Mole of 2,6-di-t-butylphenol, 1.0 mole of acetone, 0.15 mole of chloroform, 0.1 mole of N'-(2-isopropylaminoethyl)-3,3,5,5,-tetramethyl-2-piperizinone were added to a reactor and mixed by stirring while being cooled by a circulating cold bath. 0.5 mole of powdered sodium hydroxide was slowly added over a period of 1 hour. The reaction mixture was stirred at 10°C overnight. The reaction mix from the reactor was filtered. The solid residue was washed with methylene chloride and the wash added to the filtrate. The filtrate was washed with 50 ml of 4N hydrochloric acid, 50 ml of 5% sodium carbonate and was then dried over sodium sulfate. The filtrate was evaporated to dryness and the dried product was washed with hexane. The resulting amide was a white solid that had a melting point of 165-169°C and a molecular weight of 515. By elemental analysis it was determined that the amide contained 72.43% carbon (72.19% calculated), 10.26% hydrogen (calculated 10.36%) and 8.02% nitrogen (calculated 8.15%).

## EXAMPLE 3

### N-(2,2,6,6-tetramethyl-4-piperidinyl)-N'-[2-(3,5-di-t-butyl-4-hydroxyphenyl)-2-methyl propanecarboxyl]-1,6-hexanediamine

0.2 mole of 2,6-di-t-butylphenol, 1.0 mole of acetone, 0.15 mole of chloroform, 0.1 mole of 4-hexamino-2,2,6,6-tetramethyl piperidine were added to a reactor and mixed by stirring while being cooled by a circulating cold bath. 0.5 mole of powdered sodium hydroxide was slowly added in small portions over a period of 1 hour. The reaction mixture was stirred at 10°C overnight. The reaction mix from the reactor was filtered. The solid residue was washed with methylene chloride and the wash added to the filtrate. The filtrate was washed with 50 ml 4N hydrochloric acid, 50 ml of 5% sodium carbonate and was then dried over sodium sulfate. The filtrate was evaporated to dryness and the dried product was washed with hexane. The resulting amide was a white solid that had a melting point of 255-260°C and a molecular weight of 983.5. By elemental analysis it was determined that the amide contained 76.40% carbon (76.38% calculated), 10.71% hydrogen (calculated 10.90%) and 5.79% nitrogen (calculated 5.84%).

To demonstrate the stabilizing activity of the amides of this invention, test samples of the amides in polypropylene were prepared by mixing the stabilizer compounds with polypropylene in a Brabender Plasticorder fitted with a Cam-Head (mixing chamber). The polypropylene is first masticated for 1 1/2 minutes at 190°C. Then the stabilizer is added, followed by 3 minutes additional mixing. The mass is removed and pressed into 20 mil thick sheets. From these sheets are cut 1" x 1" plaques for oven aging. TypeC (3" x 1/8") tensil bars are cut for UV stability tests.

Thermal/oxidative stability (oven aging) testing consisted of aging the samples in triplicate in an air-circulating oven at 125°C. The time to catastrophic crumbling (failure) of the plaque was measured and reported as days to failure. Each sample contained 0.1 weight part of the named amide per 100 weight parts of polypropylene (pHr). The following results, in days to failure, were obtained:

| | 125°C |
|---|---|
| Control | 2 |
| N-methyl-N'-(2,2,6,6-tetramethyl-4-piperidinyl)-2-(3,5-di-t-butyl-4-hydroxyphenyl)-2-methylpropionamide | 7 |
| N-Isopropyl-N'-[2-(3,3,5,5-tetramethyl-1-piperazinyl)]ethyl-2-(3,5-di-t-butyl-4-hydroxyphenyl)-2-methylpropionamide | 9 |

Samples containing 0.1 weight part of the amide listed below was tested for ultraviolet light stability, i.e., resistance to degradation by UV radiation. The samples were tested in an Atlas Xenon Weatherometer, Model No. 65-WR, equipped with a 6500 watt Xenon burner tube in accordance with ASTM #D2565-79-A. The black panel temperature was 60°C. The samples were subjected to an 18-minute water cycle every 2 hours. The time in hours to a 50% loss in tensile strength was determined. For comparison purposes a samples with no amide was tested. The following results, in hours, were obtained:

| | Hours |
|---|---|
| Blank (Control) | 220 |
| N-Isopropyl-N'-[2-(3,3,5,5-tetramethyl-1-piperazin-2-onyl)ethyl]-2-(3,5-di-t-butyl-4-hydroxyphenyl)-2-methylpropionamide (0.1 phr) | 1280 |

The N-(2,2,6,6-tetraalkyl-4-piperidinyl)--3,5-di-alkyl-4-hydroxyphenyl)-$\alpha'$,$\alpha''$-dialkylacetamides and N-alkyl-N'-(3,3,5,5-tetraalkyl-2-keto-1-piperazin ealkyl)-$\alpha$-(3,5-di-alkyl-4-hydroxyphenyl)-$\alpha'$,$\alpha''$-dialkylacetamides as defined herein provide exceptional heat stabiity and resistance to ultraviolet degradation to polyolefin polymers. They are especially useful for the stabilization of $\alpha$-monoolefin homopolymers and copolymers, wherein the $\alpha$-monolefin contains 2 to about 8 carbon atoms. High and low-density polyethylene, isotactic and atactic polypropylene, polyisobutylene, and poly(4-methyl-1-pentene) have excellent resistance to heat and oxygen when stabilized with the combinations of the present invention. Ethylene-propylene copolymers and ethylene-propylene terpolymers, generally containing less than about 10 percent by weight of one or more monomers containing multiple unsaturatation provided, phenols; ring opened olefin polymers and the like. Polymer blends, that is, physical admixture of two or more polymers may also be stabilized in accordance with the present invention.

In addition to polymeric materials, the present compounds may stabilize a wide variety of other organic materials. Such compounds include: waxes, synthetic and petroleum-derived lubricating oils and greases; animal oils such as, for example, fat, tallow, lard, codliver oil, sperm oil; vegetable oils such as castor, linseed, peanut, palm, cotton seed, and the like; fuel oil, diesel oil, gasoline and the like.

The (3,5-dialkyl-4-hydroxyphenyl)-substituted amides may be used with other stabilizers including hindered or partially hindered phenols, hindered amine light stabilizers, phosphites, o-hydroxybenzophenones, and the like. Typical hindered phenols are the hydrox yphenylaklyleneyl isocyanurates such as the symmetrical tris(3,5-di-t-alkyl-4-hydroxybenzyl) isocyanurates; tetrakis[methylene 3-(3',5'-dialkyl-4'-hydroxyphenyl)propanoate] methanes wherein the alkyl groups contain 1 to 8 carbon atoms, such as tetrakis [methylene 3-(3',5'-di-t-butyl-4'-hydroxyphenyl)propanoate]methane; alkyl (3-3',5'di-t-butyl-4'-hydroxyphenyl)propionates wherein the alkyl groups contain 1 to 18 carbon atoms, such as octadecyl 3-(3',5'-di-t-butyl-4-hydroxyphenyl)propionate; 1,3,5-trimethyl-2,4,6-tris[3,5-dialkyl-4-hydroxybenzyl)benzene; 1,3,5-tris-(3,5-di-t-butyl-4-hydroxyhydrocinnamoylethyl)-s-triazine-2,4,6-(1H,3H,5H)-trione; 2,2'-alkylidene bis (4,6-dialkylphenol)-s wherein the alkyl group contains 1 to 8 carbon atoms, such as 2,2'-methylene bis(4,6-di-t-butylphenol), 2,2'-ethylidene bis(4,6-di-t-butylphenol), and 2,2'-methylene bis(4-methyl-6-t-butylphenol), and the like.

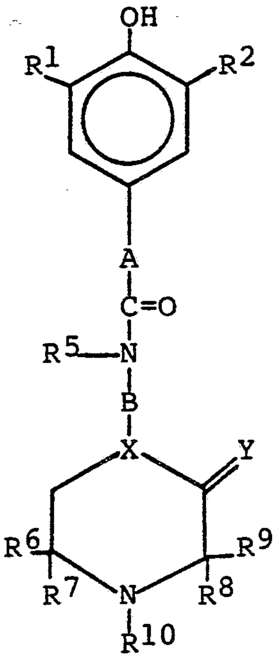
For example, the following combinations provided excellent ageing in polypropylene as shown in the Weatherometer: 0.05 phr of N-isopropyl-N'-[2-(3,3, 5,5-tetramethyl-1-piperazin-2-onyl)ethyl]-2-(3,5-di-t-butyl-4-hydroxyphenyl)-2-methylpropionamide, 0.05 phr of 1,1'-(1,2-ethanediyl)bis(3,3,5,5-tetramethyl piperazinone, and 0.05 phr of 2,2',2"-tris[3(3,5-di-t-butyl-4-hydroxyphenyl)propionyloxy]ethyl isocyanurate - more than 2,000 hours; 0.125 phr of N-isopropyl-N'-[2-(3,3, 5,5-tetramethyl-1-piperazin-2-onyl)ethyl]-2-(3,5-di-t-butyl-4-hydroxyphenyl)-2-methyl propionamide, 0.05 phr of 2,2',2"-tris[3(3,5-di-t-butyl-4-hydroxyphenyl)-propionyloxy]ethyl isocyanurate, and 0.125 phr of 3,9-bis (octadecyloxy)-2,4,8,10-tetraoxa-3,9-diphosphaspiro[5, 5]-undecane - more than 2,000 hours.

The compounds are readily incorporated into materials to be patented by dissolving or dispersing them with the materials, in liquids, dispersions, soluations, and solid forms. If the material is a solid, especially a polymeric solid such as rubber or a plastic, the compounds can be admixed using mixers such as Banburys, extruders, two-roll mills, and the like, following conventional techniques. One way to disperse the compounds in plastic materials is to dissolve or suspend the compounds in a solvent or diluent, mix the mixture with a plastic in powder or solution form, and then evaporate the solvent.

Compositions containing the novel combination of compounds can also contain other known compounding ingredients such as fillers like carbon black, silica, metal carbonates, talc, and the like; pigments and colorants; curative ingredients like sulfur and peroxides, and vulcanization accelerators; fungicides; processing aids, reinforcing agents and standard ingredients known to the art. Other ingredients known in the art as ultraviolet light, thermal and/or oxidative stabilizers can also be used.

## Claims

1. N-(substituted cyclic alkyleneimine)-α-(3,5-di-alkyl-4-hydroxyphenyl)-α',α"-dialkyl acetamides having the general formula



wherein $R^1$ and $R^2$ are hydrogen or alkyl and cycloalkyl groups containing 1 to 12 carbon atoms, alkylcycloalkyl and aryl or alkaryl groups wherein the alkyl groups contain 1 to 8 carbon atoms; $R^5$, $R^6$, $R^7$, $R^8$ and $R^9$ are hydrogen, alkyl and cycloalkyl groups containing 1 to 12 carbon atoms, alkylcycloalkyl and aryl or alkaryl groups wherein the alkyl groups contain 1 to 8 carbon atoms, A is (1) an alkylidene group of the formula

$R^3-\overset{|}{\underset{|}{C}}-R^4$ wherein $R^3$ and $R^4$

are alkyl groups containing 1 to 18 carbon atoms, $C_1$-$C_{18}$, or (2) cycloalkyl groups containing 5 to 12 carbon atoms, and alkaryl groups wherein the alkyl groups contain 1 to 8 carbon atoms; B is a bond or alkylene group -$(CH_2)_n$-wherein n is 1 to 8; $R^{10}$ is hydrogen or alkyl radicals containing 1 to 18 carbon atoms, O or OH; X is N or CH; and Y is $H_2$ or O.

2. N-(2,2,6,6-tetraalkyl-4-piperidinyl)-α-(3,5-di-alkyl-4-hydroxyphenyl)-α′,α″-dialkylacetamides and N-alkyl-N′-(3,3,5,5,-tetraalkyl-2-keto-1-piperazinyl)-α-(3,5-di-alkyl-4-hydroxyphenyl-α′,α″-dialkylacetamides of Claim 1 wherein $R^1$ contains 1 to 8 carbon atoms; $R^2$ contains 1 to 5 carbon atoms; $R^3$ and $R^4$ contain 1 to 8 carbon atoms, the aryl groups are phenyl or naphthyl; (2) is cyclohexyl, methylcyclohexyl, cycloheptyl, or dicyclohexyl, and $R^1$, $R^2$, $R^5$, $R^6$, $R^7$, $R^8$ and $R^9$ are alkyl groups containing 1 to 8 carbon atoms, and $R^{10}$ is hydrogen or an alkyl group containing 1 to 8 carbon atoms; in B n is 2 to 4; and X is CH.

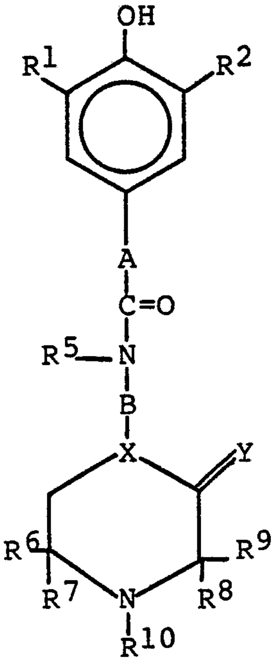
3. The acetamides of Claim 2 wherein at least one of $R^1$ and $R^2$ is t-butyl or t-amyl and $R^3$ and $R^4$ are alkyl radicals containing 1 to 4 carbon atoms.

4. N-methyl-N′-(2,2,6,6-tetramethyl-4-piper-dinyl)-2-(3,5-di-t-butyl-4-hydroxyphenyl)-2-methylpropionamide of Claim 3.

5. N-isopropyl-N′-[2-(3,3,5,5-tetramethyl-1-piperazin-2-onyl)ethyl]-2-(3,5-di-t-butyl-4-hydroxyphenyl)-2-methylpropionamide of Claim 3.

6. N-(2,2,6,6-tetramethyl-4-piperidinyl)-N′-[2-(3,5-di-t-butyl-4-hydroxyphenyl)-2-methylpropanecarboxy-1]-1,6-hexanediamine of Claim 3.

7. A composition comprising (1) organic materials subject to degradation and stabilizing amounts of (2) N-(substituted cyclic alkyleneimine)-α-(3,5-dialkyl-4-hydroxyphenyl)-α′,α″-dialkyl acetamides having the general formula



wherein $R^1$ and $R^2$ are hydrogen or alkyl and cycloalkyl groups containing 1 to 12 carbon atoms, alkylcycloalkyl and aryl or alkaryl groups wherein the alkyl groups contain 1 to 8 carbon atoms; $R^5$, $R^6$, $R^7$, $R^8$ and $R^9$ are hydrogen, alkyl and cycloalkyl groups containing 1 to 12 carbon atoms, alkylcycloalkyl and aryl or alkaryl groups wherein the alkyl groups contain 1 to 8 carbon atoms; A is (1) an alkyidene group of the formula

$R^3\text{-}\overset{|}{\underset{|}{C}}\text{-}R^4$ wherein $R^3$ and $R^4$

are alkyl groups containing 1 to 18 carbon atoms, $C_1$-$C_{18}$, or (2) a cycloalkyl group containing 5 to 12 carbon atoms, and alkaryl groups wherein the alkyl groups contain 1 to 8 carbon atoms; $R^{10}$ is hydrogen or alkyl radicals containing 1 to 18 carbon atoms, O or OH; X is N or CH; Y is $H_2$ or O; and B is a bond or alkylene group -$(CH_2)_n$-wherein n is 1 to 8.

8. Compositions of Claim 7 wherein $R^1$ contains 1 to 8 carbon atoms; $R^2$ contains 1 to 5 carbon atoms; $R^3$ and $R^4$ contain 1 to 8 carbon atoms; (2) is cyclohexyl, methylcyclohexyl, cycloheptyl, or dicyclohexyl, the aryl groups are phenyl or naphthyl; and $R^1$, $R^2$, $R^5$, $R^6$, $R^7$ and $R^8$ are alkyl groups containing 1 to 8 carbon atoms; and $R^{10}$ is hydrogen or an alkyl group containing 1 to 8 carbon atoms; in B n is 2 to 4; and X is CH.

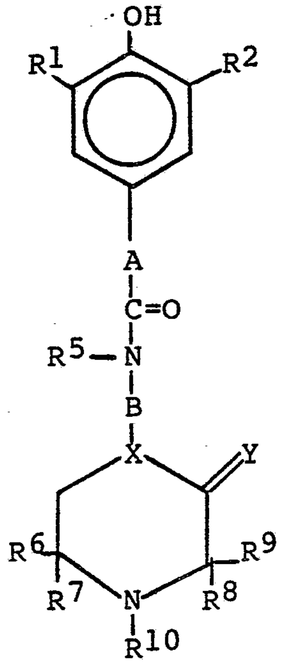
9. Compositions of Claim 8 wherein at least one of $R^1$ and $R^2$ is t-butyl or t-amyl and $R^3$ and $R^4$ are alkyl radicals containing 1 to 4 carbon atoms.

10. Compositions of Claim 9 wherein in (1) said organic material is a polymer and (2) is N-methyl-N'-(2,2,6,6-tetramethyl-4-piperidinyl)-2-(3,5-di-t-butyl-4-hydroxyphenyl)-2-methylpropionamide.

11. Compositions of Claim 9 wherein in (1) said organic material is a polymer and (2) is N-isopropyl-N'-[2-(3,3,5,5-tetramethyl-1-piperazin-2-onyl)ethyl]-2-(3,5-di-t-butyl-4-hydroxyphenyl)-2-methylpropionamide.

12. Compositions of Claim 9 wherein in (1) said organic material is a polymer and (2) is N-(2,2,6, 6-tetramethyl-4-piperidinyl)-N-2-[(3,5-di-t-butyl-4-hydroxyphenyl)-2-methylpropanecarboxyl]-1,6-hexanediamine.

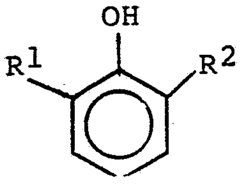
13. A process for preparing N-(substituted cyclic alkyleneimine)-α-(3,5-di-alkyl-4-hydroxyphenyl)-α,α'-dialkyl acetamides having the general formula



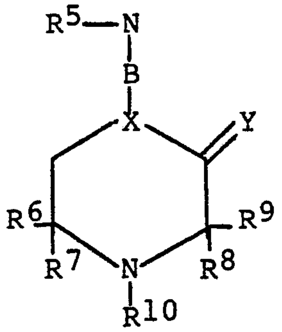
wherein $R^1$ and $R^2$ are hydrogen or alkyl and cycloalkyl groups containing 1 to 12 carbon atoms, alkylcycloalkyl and aryl or alkaryl groups wherein the alkyl groups contain 1 to 8 carbon atoms; $R^5$, $R^6$, $R^7$, $R^8$ and $R^9$ are hydrogen, alkyl and cycloalkyl groups containing 1 to 12 carbon atoms, alkylcycloalkyl and aryl or alkaryl groups wherein the alkyl groups containing 1 to 8 carbon atoms; A is (1) an alkyidene group of the formula

$R^3\text{-}\underset{|}{\overset{|}{C}}\text{-}R^4$ wherein $R^3$ and $R^4$

are alkyl groups containing 1 to 18 carbon atoms, $C_1$-$C_{18}$, or (2) a cycloalkyl group containing 5 to 12 carbon atoms, and alkaryl groups wherein the alkyl groups contain 1 to 8 carbon atoms; $R^{10}$ is hydrogen or alkyl radicals containing 1 to 18 carbon atoms, O or OH; X is N or CH, B is a bond or alkylene group $-(CH_2)_n-$ wherein n is 1 to 8; and Y is $H_2$ or O; comprising reacting together a 2,6-dialkylphenol having the formula



wherein $R^1$ and $R^2$ have the meanings above, a ketone selected from the group consisting of dialkyl ketones, cycloalakanones, alkcycloalkanones, and alkaryl ketones wherein the alkyl groups contain 1 to 18 carbon atoms, a haloform selected from the group consisting of chloroform and bromoform, and an alkali metal hydroxide and piperidines and the piperazines, having the general formula



wherein $R^5$, $R^6$, $R^7$, $R^8$ and $R^9$ are hydrogen, alkyl, cycloalkyl or alkaryl radicals containing 1 to 12 carbon atoms; $R^{10}$ is hydrogen, alkyl or arkaryı as defined herein, oxygen radical or hydroxyl; X is N or CH; in B n is 2 to 4; and y is $H_2$ or O.

14. A process of Claim 13 wherein $R^1$ contains 1 to 18 carbon atoms and $R^2$ contains 1 to 5 carbon atoms, the ketone is a dialkyl ketone of the formula

$$R^3-\overset{O}{\overset{\parallel}{C}}-R^4$$

wherein $R^3$ and $R^4$ are alkyl radicals containing 1 to 8 carbon atoms, cyclopentanone, cyclohexanone, cycloheptanone and alkyl derivatives thereof wherein the alkyl groups contain 1 to 4 carbon atoms, $R^1$, $R^2$, $R^3$, $R^4$, $R^5$, $R^6$, $R^7$, $R^8$ and $R^9$ are alkyl groups containing 1 to 8 carbon atoms, and X is CH.

European Patent
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# EUROPEAN SEARCH REPORT

Application number

EP 86 10 9748

## DOCUMENTS CONSIDERED TO BE RELEVANT

| Category | Citation of document with indication, where appropriate, of relevant passages | Relevant to claim | CLASSIFICATION OF THE APPLICATION (Int. Cl.4) |
|---|---|---|---|
| Y | EP-A-0 112 802 (CIBA-GEIGY AG)<br>* page 1, lines 7-15; table I, compound 12 * | 1-3,7-9,13,14 | C 07 D 211/26<br>C 07 D 211/74<br>C 07 D 211/58<br>C 07 D 211/94<br>C 07 D 241/04<br>C 07 D 241/08<br>C 07 D 295/22<br>C 07 D 295/12<br>C 08 K 5/34 |
| Y | EP-A-0 082 817 (CIBA-GEIGY AG)<br>* page 14, line 15 - page 15, line 5; table I, compounds 1, 29; claim 1 * | 1-3,7-9,13,14 | |
| Y | EP-A-0 141 419 (THE B.F. GOODRICH CO.)<br>* page 36, line 7 - page 44, line 2 * | 1-3,7-9,13,14 | |
| | | | **TECHNICAL FIELDS SEARCHED (Int. Cl.4)** |
| A | DE-A-2 621 870 (CIBA-GEIGY AG)<br>* page 22, lines 4, 5; claims 1-21 * | 1,7 | C 07 D 211/00<br>C 07 D 241/00<br>C 07 D 295/00<br>C 08 K 5/00 |
| A | EP-A-0 019 237 (MONTEDISON S.P.A.)<br>* claims 1, 3-15 * | 1,7 | |
| A | DE-A-2 647 452 (CIBA-GEIGY AG)<br>* claims 1, 13-24 * | 1,7 | |

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The present search report has been drawn up for all claims

| Place of search | Date of completion of the search | Examiner |
|---|---|---|
| BERLIN | 12-03-1987 | VAN AMSTERDAM L.J.P. |

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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